Fuel 214 (2018) 1–13

Contents lists available at ScienceDirect

Fuel

journal homepage: www.elsevier.com/locate/fuel

Full Length Article

Non-linear gas desorption and transport behavior in coal matrix: Experiments and numerical modeling

Peng Liu[a,b,c], Yueping Qin[a,b], Shimin Liu[c,*], Yongjiang Hao[d]

[a] College of Resources & Safety Engineering, China University of Mining & Technology (Beijing), Beijing 100083, China
[b] State Key Laboratory of Coal Resources & Safe Mining, CUMTB, Beijing 100083, China
[c] Department of Energy and Mineral Engineering, G^3 Center and EMS Energy Institute, Pennsylvania State University, University Park, PA 16802, USA
[d] College of Environment and Safety, Taiyuan Universtiy of Science and Technology, Taiyuan 030024, China

ARTICLE INFO

Keywords:
Non-linear desorption
Gas transport in coal matrix
Density
Fick
Experiment
Modeling

ABSTRACT

Gas desorption and transport in coal matrix plays pivot roles to estimate *in situ* gas content, forecast gas production from coalbed methane (CBM) wellbores, classify the gas/coal outburst proneness of coal seams and estimate gas emission rate for active mine ventilation planning. Only using Fick's law to depict methane transport in coal matrix may result in an erroneous prediction because it uses only adsorbed phase gas to calculate methane concentration gradient. In this study, a series of coal-methane ad/desorption experiments were carried out under different pressure boundary conditions. Following this, an effort is made to propose a semi-empirical desorption model describing the entire methane diffusion process and discuss its superiority and applicability by comparing to various commonly used models. The proposed approach includes two different theoretical models (Fick diffusion model, assuming concentration-difference transports gas; and Density model, assuming density-difference transports gas), to model methane diffusion corresponding to the experimental sections conducted in this study. Afterward a series of comparisons between the experimental desorption data and two sets of simulated desorption data obtained by numerically calculating the two theoretical models were conducted, and it shows that Density model exhibited a higher accuracy over Fick model. The proposed Density model is more effective in describing the non-linear gas diffusion behavior in coal matrix for the experimentally studied coals. Essentially, the Density model covers and promotes the Fick diffusion model, and is competent in mathematically modeling both adsorbing gas and non-adsorbing gas transport behavior in porous media. Moreover, the Density model can be directly incorporated to the existing dual-porosity model to model methane migration in coal matrix in coal seam.

1. Introduction

Coalbed methane (CBM) is known as miners' curse and mine explosion is one of the main coal mine disasters in coal mining history. In recently years, CBM has emerged as one of the clean natural gas resource due to the successful extraction from both virgin coal and active coal mines. Methane is also known as a stronger greenhouse gas, 21 times more potent than CO_2 in terms of contributing to global warming [1–4]. It is practically important to study methane transport behavior in coal since it directly relates to: (*a*) determining gas content and gas storage capacity [5]; (*b*) predicting methane emissions [6,7]; (*c*) evaluating coal-gas outburst prone potential [8]; (*d*) improving efficiency of gas extraction [9], and (*e*) secondary enhanced-CBM recovery [10,11], *etc.*

Significant efforts have been made to understand and characterize methane transport in coal matrix. As a means to describe gas diffusion process and methane emission behavior, various models both theoretical and empirical ones, have been proposed and studied for different coals. Based on investigations of gas diffusion in zeolite sand, Barrer proposed the classical diffusion model (unipore model) and a simplified mathematical formula to estimate diffusion rate [12]. Nandi and Walker studied coal-methane diffusion behavior and determined the diffusion coefficient for the early stage of desorption process according to the classical diffusion model [13,14]. Yang et al. derived the analytical solution of the classical model, and compared theoretical estimated results with experimental data and found that the two are roughly consistent [15]. Smith et al. found a relative large discrepancy between theoretically estimated results and experimentally measured data for the late stage of diffusion process [6]. Ruckenstein et al. proposed a bidisperse diffusion model to better describe the diffusion

---

[*] Corresponding author.
E-mail address: szl3@psu.edu (S. Liu).

http://dx.doi.org/10.1016/j.fuel.2017.10.120
Received 28 July 2017; Received in revised form 19 October 2017; Accepted 24 October 2017
0016-2361/

process for bi-model coal pore structures and it was found that the accuracy was improved by using bidisperse model [16]. Both Clarkson et al. and Shi et al. presented an improved bidisperse diffusion model to fit the experimental data of methane diffusion in bituminous coal [10,17]. A simplified bidisperse diffusion model was proposed to reduce the computational complexity of the bidisperse model [9]. A Fickian diffusion-relaxation model that split the diffusion process into a primary and secondary stage was proposed in the light of the bidisperse model [18]. Besides, some scholars introduced time-dependent or pressure/concentration-dependent diffusion coefficient and combined it with Fickian diffusion model to depict entire timescale desorption process to acquire a good fit with the experimental desorption data [19–23]. All mentioned models assumed that methane flow is concentration gradient driven transport and Fick's Law is valid for modeling gas diffusion through coal matrix.

The pore structure of coal matrix is complex and its size ranges from angstrom (Å) to micrometers (μm) [24–28]. Because of this wide pore size range, it is suspected that the gas transport in coal matrix only involves Fick's mass influx. Some scholars believe the gas transport in coal matrix is a multi-mechanism process. Alley argued that applying only Darcy's law can be practically effective to describe methane transport in lump coals unless the coals have been subjected to excessive damages [29]. Shi et al. believed methane emission from coal matrix is a combination of methane diffusion and methane seepage, which one dominates the whole process of matrix gas release depends on the specific coal pore structure [30]. Laboratory tests revealed that the existence of two types of pores with coal matrix, a diffusion pore controlling methane desorption and diffusion, and a permeation pore dominating methane permeation [24,31,32]. The triple porosity/dual permeability model, which assumes methane migration *via* desorption and diffusion from micro-pores into meso/macro-pores and then followed by the transports *via* Darcy flow within meso/macro-pores and fractures, exhibits a better performance than the dual porosity/single permeability model in terms of simulating CBM recovery [31–34]. Qin et al. found the experimental data of coal-methane ad-/de-sorption matches well with the simulated results of modeling methane emissions from spherical coal particles with employing Darcy's law, and suggested that Darcy's law can be applied to describe methane migration in coal matrix [35,36].

Besides these theoretical models aforementioned, empirical models, as an easy, painless and rapid method to calculate gas desorption, also were used in mining industry for field screening applications. So far, a number of empirical models, such as Bolt model [37], Airey model [29], ВСТИНОВ model [38], were proposed based on experimental data or field data regression. These empirical models were used to estimate desorption amount or rate. Some models can accurately describe the initial stage of desorption process, but fail to define in the whole desorption process. Others can predict the final desorption amount, but fail to match the desorption trend during desorption process. So few models can be used for the entire desorption process. Moreover, most empirical models are proposed for a single type of coal and their extension for other coal application is hard to justify.

Although the gas transport mechanism inside coal matrix has been studied over a few decades, the fundamental mechanism still needs further discussion and a uniform applicable methane transport framework is required for field application. This study describes a series of experiments on methane ad/desorption on coal matrix and conducts a succession of simulations on coal-methane diffusion. This study aims at making a contribution to the methane diffusion mechanisms and mathematical description of coal-gas diffusion process in coal matrix.

2. Experimental

2.1. Coal sampling and preparation

Table 1
Number and particle sizes (diameter) of coal samples.

Sample	Number	Particle size range (μm)	Average particle size (μm)
YC sample	YC1	4000–4750	4359
	YC2	1000–1180	1090
	YC3	425–550	487.5
	YC4	250–270	260
XW sample	XW1	42834–42967	42946
	XW2	11600–13800	12760
	XW3	3350–4000	3675
	XW4	1180–1400	1290

Two coals were used in the experiments: a bituminous coal from Yangcao coal mine in Northeast China and a sub-bituminous coal from Xuanwei coal mine in the Southwest china. These two coal samples were pulverized and sieved to the desired particle size, and the distribution intervals are shown in Table 1. In order to determine the average size for samples with lager particle size such as YC1, XW1 and XW2, the size of coal particles in three different directions are first measured, and the average of the three value of length is considered as the diameter of the particle, and then similarly we measured the particle diameter of 20 coal particles from the same coal sample and computed the average value of 20 particle diameters, the average value was taken as the average particle size of this coal sample. For the particle size of the remaining samples were determined by averaging the upper and lower limits of the size range. The average particle size of all used samples is also listed in Table 1. For ad/desorption measurements on dry coal, the coal samples were dried in the oven at 373 K for 24 h, which is a common approach to remove the moisture of coal in the published literatures [7,10,39,40].

2.2. Experimental apparatus and procedures

The experimental apparatus used in this work includes the ad-/desorption system, the temperature control system and the data acquisition system (DAS). The schematic of the experiment apparatus is shown in Fig. 1. The ad-/de-sorption system mainly consists of a stainless-steel reference tank, a stainless-steel sample tank, gas cylinders and connecting tube. To ensure a constant temperature in all experiments, the tanks were placed into the isothermal oven within 0.1 K. Two high-precision pressure transmitters are connected to the DAS to monitor the pressure change in the sample tank and reference tank, and the pressure data in every second was recorded during the tests.

Before starting the desorption process, the coal sample was initially saturated with methane and waits until the adsorption equilibrium in the sample tank for given pressure of methane. The equilibrium pressure was monitored and recorded. And this pressure was termed as the initial pressure for the desorption experiment. In this study four different initial pressures were used, 0.5, 1, 2, 4 MPa, respectively. The gas desorption was carried out under two different pressure boundary conditions. One is constant atmospheric pressure boundary condition,

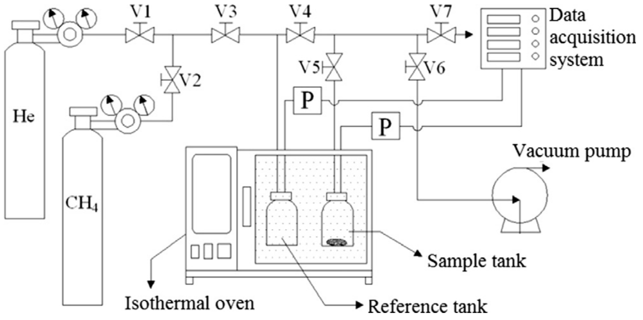

Fig. 1. Schematic of experimental apparatus used for the ad/desorption experiments (p stands for pressure gauge; V stands for needle valve).